

# Thermodynamics of a general stochastic model of magnetic hysteresis

D. M. Clatterbuck and J. W. Morris, Jr.

*Lawrence Berkeley National Lab and Department of Materials Science and Engineering, University of California, Berkeley, CA 94720*

The thermodynamics of a general stochastic model of magnetic hysteresis are analyzed and the implications on past and future work on hysteresis models are discussed. The conditions of equilibrium and stability are derived for a random potential model with no assumptions about the form of the potential. The analysis demonstrates that the average over all instances of the potential is related to the equilibrium behavior of the system and the deviations of each instance from this average is related to the domain wall pinning properties. This allows one to analyze the ad-hoc assumptions made in earlier models of magnetic hysteresis and the Barkhausen effect. It also suggests a clear starting point for future work on random potential models.

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## I. Introduction

The complexity of the magnetization processes that occur in ferromagnets has led many investigators to search for simplified approaches to allow for the interpretation of hysteresis behavior. In the history of the study of hysteresis, a prominent role has been played by the type of model first proposed by Néel<sup>1</sup>, which is sometime referred to as a random potential model. In this model, the system is represented by a single degree of freedom which evolves in a random potential energy landscape. This potential energy function contains multiple metastable states, causing the system to exhibit hysteresis when subjected to an applied magnetic field. Similar types of metastable states are created in ferromagnetic materials when the domain walls become pinned at defects.

The random potential model was first proposed by Néel as an explanation for the low field Rayleigh region encountered in many materials. This type of model was further studied by Pfeiffer<sup>2</sup> as well as Kronmuller and co-workers in a number of papers<sup>3,4,5</sup>. More recently, Bertotti and co-workers have explored the use of stochastic differential equations for representing the random potential<sup>6,7,8</sup>. One difficulty in the use of random potential models has been the need for ad-hoc assumptions about the form of the effective potential. Previous investigators have not carried out a thermodynamic analysis of a generalized random potential model; therefore, is instructive to consider what limits are placed on the potential by the thermodynamics of the model and if any further insight can be obtained from such an analysis.

## II. Thermodynamic Analysis

The functional integration approach to hysteresis has recently been proposed by Bertotti, Mayergoyz, Basso, and Magni<sup>9</sup>. It is a general mathematical description of hysteresis which is in a sense a generalization of the approach developed by Néel. While the functional integration approach has been presented in a general way for any two conjugate work variables  $H$  and  $X$ , let us consider the specific case where  $H$  represents the magnetic field and  $X$  represents the magnetization  $M$ .

Let us assume that the magnetization process can be reduced to a single dominant degree of freedom (i.e.  $M$ ), such that we don't need to know spatial variation of the orientation of all the moments in the material, only the average value over an entire bulk sample in order to identify the state of the system. Only a single component of the magnetization in a direction parallel to the magnetic field will be considered to simplify the problem to a scalar model. Now let the Helmholtz free energy of the system be given as a function of  $M$  by  $F(M)$ . We will consider isothermal processes and omit the variable  $T$  for conciseness of notation. Because of the presence of structural disorder in the system, we expect that  $F(M)$  will contain multiple metastable states which give rise to the non-equilibrium hysteresis behavior of the system. Let the relevant free energy of the system  $G_L$  be given by  $G_L(M;H) = F(M) - \mu_0 M H$ , where the subscript L indicates that we are considering the Landau free energy or the free energy of the system as a function of  $H$  in the case where  $M$  is constrained away from its equilibrium value.

The functional integration approach considers that the system contains a set of generating functions  $\mathbf{g}$  which describe the free energy profile,  $\mathbf{g} = dF/dM$ . For each generating function, one determines the evolution of the system by considering the action of a magnetic field on some initial state which brings the system to the final state through a envelope-like construction as shown in Figure 1. As the magnetic field is varied, the system remains in local equilibrium ( $\mu_0 H = dF/dM$ ) until it reaches a maximum or minimum in the  $dF/dM$  curve and then proceeds by a spontaneous jump to the next available local minimum (which precludes the possibility of inertial effects which may allow the system to reach more distant minima). Of particular interest is the case where  $\mathbf{g}$  are statistically independent. This allows the hysteresis behavior of a system of generating functions to be found from a statistical average of these trajectories weighted by their probability of occurrence,  $p$ .

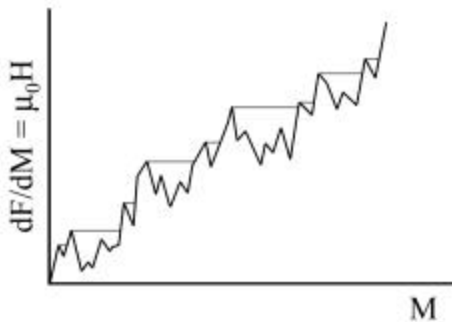


Figure 1. Generating function and envelope operation which define the evolution of the system.

Let us now consider some of the consequences of the thermodynamics of a system represented by a single generating function  $g$ . The condition of local equilibrium is that a small change in state must increase the free energy,  $dG_L \geq 0$ . At constant magnetic field this can be written as:

$$dG_L = \left[ \left( \frac{\partial G_L}{\partial M} \right)_H \right] dM = \left[ \left( \frac{\partial F}{\partial M} \right)_H - m_0 H \right] dM = [g(M) - m_0 H] dM \geq 0. \quad (1)$$

Since  $\delta M$  can be positive or negative this means that we must have

$$g(M) = m_0 H, \text{ or } \left( \frac{\partial F}{\partial M} \right)_H = m_0 H, \quad (2)$$

which expresses the condition of equilibrium in terms of either  $g$  or  $dF/dM$ .

The condition of local stability is that the second variation of the free energy must be positive,  $d^2 G_L > 0$ . We can write this as:

$$dG_L = \left[ \left( \frac{\partial F}{\partial M} \right)_H - m_0 H \right] dM = [g(M) - m_0 H] dM$$

$$d^2 G_L = \frac{\partial g}{\partial M} dM^2 > 0, \text{ thus } \frac{\partial g}{\partial M} > 0; \left( \frac{\partial^2 F}{\partial M^2} \right)_H > 0 \quad (3)$$

We see that the condition of stability requires that the generating function be an increasing function for either increasing or decreasing magnetic field histories. Thus portions of the generating function with negative slope are unstable. The conditions of local equilibrium and stability are consistent with the proposed description of the system in that the system follows the condition of local equilibrium up to a maximum (or down to a minimum) in the generating function at which point the system becomes unstable owing to the fact that the generating function is no longer an increasing function and then proceeds to the next available position of local equilibrium.

Under constant  $H$  and  $T$ , a decrease in  $G_L$  corresponds to the dissipation of energy into the thermal reservoir. We can find this dissipated energy from:

$$\Delta G_L = G_L(M_2; H) - G_L(M_1; H) = \int_{M_1}^{M_2} \left( \frac{\partial G_L}{\partial M} \right)_H dM; \quad \Delta G_L = \int_{M_1}^{M_2} \left( \frac{\partial F}{\partial M} \right)_H - m_0 H dM. \quad (4)$$

From equation 4 we see that the energy dissipated is clearly the difference between the energy stored represented by the first term on the right and the energy supplied to the system represented by the second term on the right. The energy dissipation has a clear graphic representation as indicated by the shaded area in Figure 2. It is clear from this that for a closed loop the energy dissipation will be equivalent to the area of the loop as one would expect.

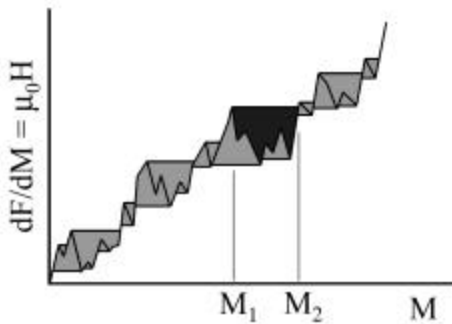


Figure 2. Graphical interpretation of energy losses. Hysteresis loss on moving from  $M_1$  to  $M_2$  is represented by dark shading. The loss associated with an entire loop is given by light shading.

We can also see from equation 4 that if a trajectory always satisfies the local condition of equilibrium there will be no energy dissipation. And thus systems which contain no unstable portions (with  $dg/dM < 0$ ) will not exhibit hysteresis. Application of the second law of thermodynamics and use of equation 4 also shows that for increasing field, a trajectory must lie on or above the generating function ( $\mathbf{m}_b H \geq dF/dM$ ) in order to avoid a spontaneous increase in the free energy of the system in violation of the second law. For decreases in  $M$ , the trajectory must lie below the generating function.

Let us now consider a collection of independent generating functions. The energy stored in the system can be found from:

$$\Delta F = \int_{\mathbf{p}} \left[ \int_{M_1}^{M_2} \mathbf{g}(M) dM \right] dp(\mathbf{g}) = \int_{M_1}^{M_2} \left[ \int_{\mathbf{p}} \mathbf{g}(M) dp(\mathbf{g}) \right] dM ; \Delta F = \int_{M_1}^{M_2} \langle \mathbf{g}(M) \rangle dM \quad (5)$$

We see that the energy stored is simply related to the expectation value of the generating function. Thus if the trajectory follows  $\mathbf{m}_b H = \langle \mathbf{g}(M) \rangle$ , no energy dissipation (no hysteresis) will occur.

We have considered thus far situations where the system is trapped in states of local equilibrium and can not escape from these metastable states. One would expect that with sufficient thermal excitation, fluctuations may allow the system to reach lower energy states. In this respect we would like to find the global conditions of equilibrium at fixed  $H$ . At global equilibrium, any change of state increases the free energy  $G_L$ ;  $\Delta G_L > 0$ . Consider the change in free energy from the equilibrium state given by  $M_{eq}$  to some arbitrary state  $M$ ,

$$\begin{aligned} \Delta G_L &= \int_{\mathbf{p}} \left[ \int_{M_{eq}}^M \left( \frac{\partial F}{\partial M} \right)_H - \mathbf{m}_b H dM \right] dp(\mathbf{g}) = \int_{M_{eq}}^M \left[ \int_{\mathbf{p}} \left( \frac{\partial F}{\partial M} \right)_H - \mathbf{m}_b H dp(\mathbf{g}) \right] dM > 0 \\ \Delta G_L &= \int_{M_{eq}}^M \left[ \int_{\mathbf{p}} \left( \frac{\partial F}{\partial M} \right)_H dp(\mathbf{g}) - \mathbf{m}_b H \int_{\mathbf{p}} dp(\mathbf{g}) \right] dM = \int_{M_{eq}}^M [\langle \mathbf{g}(M) \rangle - \mathbf{m}_b H] dM > 0 \end{aligned} \quad (6)$$

This result has a simple graphical interpretation in the case where  $\langle \mathbf{g}(M) \rangle$  is a monotonic increasing function. The above criterion is equivalent to saying that the area between the generating function and the line  $\mathbf{m}_b H$  must be positive (above  $\mathbf{m}_b H$ ) for increases in  $M$  and negative for decreases in  $M$ . Since either increases or decreases in  $M$  are admissible we see from Figure 3 that the condition of global equilibrium is such that  $\langle \mathbf{g}(M_{eq}) \rangle = \mathbf{m}_b H$ . When  $\langle \mathbf{g}(M) \rangle$  is not a monotonic increasing function but contains a large scale instability the condition of global equilibrium is such that the net area between the generating function and the line  $\mathbf{m}_b H$  must be positive for all  $M > M_{eq}$  and negative for all  $M < M_{eq}$ .

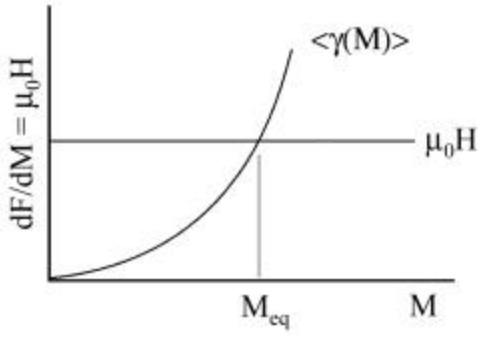


Figure 3. Graphical representation of the condition of global equilibrium.

### III. Discussion

Since the generating function is a derivative of the free energy  $F$  with respect to magnetization  $M$ , it is equivalent to a magnetic field,  $g(M) = dF/dM = \mathbf{m}_b H_F$ . Since the absolute value of  $F$  can be set to zero at some arbitrary reference, the free energy can be described entirely by an effective field  $H_F$ . From the above analysis we also see that the energy stored in the system can be related to the average value of this field which we will call  $H_m = \langle H_F \rangle$ . The difference of each free energy curve from equilibrium can be represented by  $H_p = H_F - H_m = H_F - \langle H_F \rangle$ . It is clear from this that  $\langle H_p \rangle = 0$ .

The series of points  $H_m(M)$  represents the equilibrium magnetization curve or equivalently the anhysteretic magnetization curve. Thus we expect that  $H_m$  will contain certain features consistent with known experimental evidence. Because of the symmetry of magnetic materials  $H_m(M)$  will be an odd function. It will also generally be an increasing function with 2 vertical asymptotes which correspond to magnetic saturation. One also expects that  $H_p$  will depend on the characteristics of the pinning sites and thus the properties of  $H_p$  will depend on  $M$ . For example, since the behavior of magnetic materials at saturation is reversible, we expect that  $H_p$  will tend to zero at large magnetization.

The fact that the generating potential can be subdivided into two parts based on a thermodynamic argument brings some clarity to past work on random potential models. Previous investigations have required ad-hoc assumptions about the form of the random potential in order to study the resulting hysteresis behavior. For example, Néel<sup>1</sup> assumed that the slopes of the potential (which have been represented here by  $dF/dM = \mathbf{m}_b H_F$ ) are random with a Gaussian distribution and mean of zero. From the above analysis we see that this implies that  $H_m$  is zero and  $H_F = H_p$ . In terms of physical quantities, Néel's assumption requires that the material have an infinitely steep equilibrium magnetization curve, or that  $H_m$  is not significant compared to  $H_p$ , which is a reasonable assumption in the Rayleigh region.

In the more recent work of Bertotti and co-workers on the Barkhausen effect<sup>6</sup> and magnetic hysteresis<sup>7,8</sup>, they assume that the system free energy can be separated into two distinct contributions: a random pinning term  $H_p$  with average value zero which represents the structural disorder of the system, and a large scale contribution  $H_m$  due to magnetostatic contributions which is assumed linear in  $M$  ( $H_m = M/c$ , where  $c$  is the magnetic susceptibility). In fact, the above analysis shows that this is not an arbitrary assumption and that the separation of the free energy into two contributions is a natural consequence which follows from the thermodynamics. We also see that  $H_m$  is more accurately written in terms of the equilibrium susceptibility ( $H_m = M/c_{eq}$ ). This is not inconsistent with the original Barkhausen model put forth however, because that model was restricted to the case where the domain wall dynamics are a stationary stochastic process and this requires the equivalence of the susceptibility and the equilibrium susceptibility.

The separation of the potential into two components can also act as a starting point for future developments of random potential models. One benefit is the fact that one part of the potential is simply the equilibrium or anhysteretic magnetization relationship, which can be experimentally measured. This allows for the possibility of exploring the equilibrium and pinning aspects separately which may help to simplify the problem.

Another example of the usefulness of the above analysis can be found in the low field region. In that case, one can assume that  $H_m$  will not be significant compared to  $H_p$  and that the pinning properties will be essentially constant suggesting that  $H_f$  will be a stationary stochastic process with a mean of zero. This is a clear starting point for investigation of the hysteresis properties in this regime, as one can now vary the form of the potential within the limits of this constraint and study the resulting hysteresis behavior. For example, earlier random potential models of magnetization in the Rayleigh region predicted certain scaling relations between the magnetic parameters, such as the fact that the product of the initial permeability and the coercivity are a constant. However, experiments have shown that these scaling relations do not necessarily hold in real materials<sup>10 11</sup>. Since the scaling relations are the result of certain assumptions about the effective potential, one may gain an understanding of the breakdown in these scaling relations by relaxing some of the earlier assumptions while remaining confident that the above thermodynamic analysis provides some guidance to the form of the random potential.

A thermodynamic analysis of a generalized random potential model has shown the effective potential to be composed of two terms, one related to the equilibrium behavior and the other related to the domain wall pinning in the material. This provides insight into earlier ad-hoc assumptions about the form of the potential. It also suggests a starting point for future work on these types of hysteresis models.

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- <sup>1</sup> L. Neel, *Cah. Phys.* **12**, 1 (1942).
  - <sup>2</sup> K. H. Pfeffer, *Phys. Status Solidi* **21**, 857 (1967).
  - <sup>3</sup> H. Kronmuller, *Z. Angew. Physik* **30**, 9 (1970).
  - <sup>4</sup> H. R. Hilzinger and H. Kronmuller, *J. Magn. Magn. Mater.* **2**, 11 (1976).
  - <sup>5</sup> H. Kronmuller and T. Reininger, *J. Magn. Magn. Mater.* **112**, 1 (1992).
  - <sup>6</sup> B. Allesandro, C. Beatrice, G. Bertotti, and A. Montorsi, *J. Appl. Phys.* **68**, 2901 (1990).
  - <sup>7</sup> G. Bertotti, *Hysteresis in Magnetism* (Academic Press, New York, 1998).
  - <sup>8</sup> A. Magni, C. Beatrice, G. Durin, and G. Bertotti, *J. Appl. Phys.* **86**, 3253 (1999).
  - <sup>9</sup> G. Bertotti, I. D. Mayergoyz, V. Basso, and A. Magni, *Phys. Rev. E* **60**, 1428 (1999).
  - <sup>10</sup> B. Astie, J. Degauque, J. L. Porteseil, and R. Vergne, *J. Magn. Magn. Mater.* **28**, 149 (1982).
  - <sup>11</sup> D. C. Jiles, T. T. Chang, D. R. Hougen, and R. Ranjan, *J. Appl. Phys.* **64**, 3620 (1988).